

Algorithmic Approach to Optimizing Individual Nanoparticle Control for Complex Manipulation

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Across multiple scientific fields in medicine, research, and technology, nanoparticles present various opportunities for control and influence at a scale not seen before . While the nanoscale provides a potential solution to centuries-old problems, it has its own challenges. One such challenge is the ability to complete complex actions using nanoparticles due to the lack of control over individual particles . This paper presents an algorithm that uses the optical cross-sections of nanoparticles across a range of frequencies to determine a set of nanoparticle-frequency pairings that maximizes independent control over nanoparticles. Maximized independent control not only could further enhance the performance of tasks currently being performed but also open up exciting possibilities in drug delivery and nanoelectronics.

Introduction

Achieving independent control over nanoparticles would be transformative for complex nanoscale tasks because it enables precise manipulation with complex functionality in various applications, such as medical diagnostics, drug delivery, and electronics . Achieving such control is challenging due to factors like thermal restrictions, physical stability, and interference from unintended interactions . While current methods of nanoscale manipulation often focus on the manipulation of a single object , this paper proposes an optimization algorithm for multiple nanoparticles through quantifying nanoparticle combinations and their respective frequencies through a scoring system. This scoring system evaluates optical cross-sections caused by frequencies within a specified range, aiming to maximize intended interactions and minimize unintended ones. The proposed algorithm provides greater independent control by identifying the optimal set of nanoparticle-frequency pairings that ensure selective activation and minimal crosstalk, thus improving the overall performance and precision of nanoscale tasks.

Current methods of nanoparticle control include magnetic, chemical, and mechanical methods. Magnetic methods use external magnetic fields to manipulate magnetic nanoparticles but often suffer from complexity and limited efficiency. Mechanical methods, such as acoustic tweezers, can precisely position nanoparticles but are limited by thermal effects and intricate setups, as well as the lack of complex capabilities. Chemical methods are perhaps the most promising of the bunch, allowing for precise and well-documented interactions at the chemical level, however, chemical methods are limited to the confines of chemistry .

Electromagnetic interaction with nanoparticles through localized surface plasmon (LSP) excitations offers an auspicious way of controlling nanoparticles through light-matter

interactions . LSP excitations occur when conduction electrons on the nanoparticle surface resonate with incident electromagnetic waves, leading to strong local electromagnetic fields. This interaction is pivotal for achieving precise nanoparticle control because, one, the excitation is non-propagating, and two, it allows for fine-tuning of optical properties based on the nanoparticle's material, size, and shape, among other variables .

Methods

The aim of this study was twofold. One, to demonstrate an optimization algorithm that identifies combinations of nanoparticle-frequency pairings to enhance independent control by maximizing intended interactions and minimizing unintended ones through the analysis of optical cross-sections. Two, to demonstrate and validate an optimization algorithm to improve independent control of nanoparticles through frequency-specific interactions.

Optical Cross-sections Calculation

To acquire the optical cross sections, one can either use experimental means, such as a sensitive spatial modulation technique combined with a common-path interferometer , or theoretical means, namely Mie Theory Scattering and Discrete Dipole Approximation.

Mie Scattering

Calculating Mie Scattering requires several inputs: the radius of the sphere, the wavelength of the incident electromagnetic wave, the refractive index of the sphere relative to the surrounding medium, and the number of angles for which to calculate the scattering.

The first step involves solving Maxwell's equations to describe the interaction between the electromagnetic wave and the spherical particle. These equations, expressed in spherical coordinates, govern the behavior of the electric and magnetic fields in the absence of free charges and currents. Boundary conditions at the sphere's surface and infinity ensure the solutions are physically meaningful. Next, the fields are expanded in a series of spherical harmonics, allowing for variables to be separated. The electric and magnetic fields are represented as sums of these harmonics, and the solutions involve spherical Bessel functions and spherical Hankel functions. The spherical Bessel functions describe the internal field within the sphere, while the spherical Hankel functions represent the scattered field outside the sphere. The boundary conditions are then applied at the sphere's surface, ensuring the tangential components of the electric and magnetic fields are continuous. This leads to the determination of the Mie coefficients, which characterize the scattering and absorption of light by the sphere. These coefficients depend on the sphere's size, refractive index, and the properties of the surrounding medium. The Mie coefficients are subsequently used to compute the scattering, absorption, and extinction efficiencies. Scattering efficiency quantifies the fraction of incident light that is scattered, absorption efficiency describes the fraction absorbed by the particle, and extinction efficiency accounts for the total light removed

from the incident beam (scattering plus absorption). As a final output, the optical cross-section is obtained, which encapsulates the scattering and absorption behavior of the spherical particle and provides a comprehensive measure of its interaction with the incident light. In addition to the Mie Theory, Rayleigh Scattering offers a more accurate approximation depending on the particle's diameter ratio to the incident light's wavelength.

Discrete Dipole Approximation

Alternatively, if one wants to run this algorithm using non-spherical shapes, the discrete dipole approximation method (DDA) offers a suitable theoretical approach. The DDA method involves discretizing a target particle into an array of small polarizable points or dipoles that interact with the electromagnetic field. Each dipole, whose polarizability is determined by the particle's material properties and the wavelength of light, has a dipole moment induced by the interacting light. The interactions among these dipoles are computed by solving Maxwell's equations for the dipole array, resulting in a system of coupled equations that describe the particle's electromagnetic response. The electric fields are iteratively solved until a consistent solution is achieved. Finally, the scattering and absorption cross-sections, which indicate how much light is scattered and absorbed by the particle, are calculated from the determined fields.

Algorithm

The goal of the optimization algorithm for this paper is to output a combination of n nanoparticle-frequency pairings that maximize independent control of each nanoparticle (through their paired frequency) by using the extinction, absorption, and scattering cross-sections caused by the frequencies in a range of f_1 to f_2 for all nanoparticles in a dataset. This maximizes intended interactions and minimizes unintended interactions, with intended interactions being interactions within np-freq (nanoparticle and frequency) pairings.

This algorithm can accommodate any number of added layers, sizes, and materials. Furthermore, the range of the electromagnetic wave and the step size can be adjusted, barring the addition/removal of physical properties.

Let us simplify the parameters to determine a method of maximizing independent control. Instead of looking at multiple optical cross-sections, we will only look at one. Rather than trying to optimize for n different np-freq pairings, we will optimize for only two. Consider a 5×5 table in which the vertical side contains different frequencies while the horizontal side contains different nanoparticles. Where any two of these values meet, a value for the extinction cross-section is yielded. For simplicity's sake, let's assume that the extinction cross-section directly correlates to the strength of the effect of a frequency on a nanoparticle. A table for that would look something like this:

For the following, the values for the optical cross-sections were generated by adapting the original Bohren and Huffman code for Mie Scattering. The wavelength of an electromagnetic wave with a frequency will be used during the examples for ease of use. The values were all multiplied by the most commonly used power of 10 and then rounded to the nearest half-integer value. When implementing this formula for real-life

purposes, accounting for the error derived from using floats to represent real numbers is important.

Wavelength (nanometers)	Au	Ag	Al	Cu	Ni
400	2.5	4.5	3	3	3
500	3	6	2	3.5	3
600	5.5	2.5	1	4.5	2
700	2.5	1	0.5	1	1.5
800	2	0.5	0.5	0.5	1

Remembering that the final goal for our optimization algorithm is to maximize independent control, our algorithm should find the two np-freq pairings in which the difference between the extinction cross section for a frequency intended nanoparticle and its non-pairing particle is the highest. Moreover, this must be true for the other frequencies as well. An equation depicting this situation can be denoted as the following, where S is equal to the overall score of each combination, and the optimal combination has the highest S score.

$$S = (\sigma_{ext}(np_a, f_a) - \sigma_{ext}(np_b, f_a)) + (\sigma_{ext}(np_b, f_b) - \sigma_{ext}(np_a, f_b))$$

Where:

- S : The total S-score representing independent control over the two nanoparticles.
- $\sigma_{ext}(np_a, f_a)$: The extinction cross-section for nanoparticle np_a at frequency f_a .
- $\sigma_{ext}(np_b, f_a)$: The maximum extinction cross-section for all other nanoparticles (not np_a) at the frequency f_a . This is the unintended interaction to minimize.
- The second term follows similarly for the second nanoparticle-frequency pairing np_b at frequency f_b .

Based on the above algorithm, it would output the following combination, Ag at 500 nm and Au at 600 nm, which yielded a total S score of 3.5.

From there, getting to the final proposed algorithm in this paper is simply a matter of changing the S score formula to account for any integer value n pairings, in the end, combining and splitting the extinction cross section into its absorption and scattering cross sections, as well as adding a variable W for both cross sections Weight, which can

be adjusted depending on the situation to consider one cross-section more heavily or less. This more generalized formula for the S score can be denoted as the following:

$$S = \sum_{i=1}^n \left[W_{sca} \cdot \sigma_{sca}(np_i, \lambda_i) + W_{abs} \cdot \sigma_{abs}(np_i, \lambda_i) - \sum_{j \neq i} \left(W_{sca} \cdot \sigma_{sca}(np_j, \lambda_i) + W_{abs} \cdot \sigma_{abs}(np_j, \lambda_i) \right) \right]$$

- S : The total S-score representing independent control over the nanoparticles-frequency pairings.
- W_{sca} : The scattering weight.
- $\sigma_{sca}(np_i, \lambda_i)$: The scattering cross-section for nanoparticle np_i at wavelength λ_i .
- W_{abs} : The absorption weight.
- $\sigma_{abs}(np_i, \lambda_i)$: The absorption cross-section for nanoparticle np_i at wavelength λ_i .
- $\sum_{j \neq i} \left(W_{sca} \cdot \sigma_{sca}(np_j, \lambda_i) + W_{abs} \cdot \sigma_{abs}(np_j, \lambda_i) \right)$: The undesired sum of scattering and absorption cross-sections for other nanoparticles np_j at the wavelength λ_i .

By iterating through all unique np-freq sets of n nanoparticles with this formula, the set with the highest total S-score is the most optimal set, maximizing independent control of each nanoparticle (through their paired frequency) by using the absorption, and scattering cross-sections caused by the frequencies in a range $f1$ to $f2$ for all nanoparticles in a dataset, maximizing intended interactions and minimizing unintended interactions, with intended interactions being interactions within np-freq (nanoparticle and frequency) pairings.

Example

To see how this algorithm works in complete form, one first must consider a scenario of using sequentially activated nanoparticles for localized chemotherapy . In such a scenario, both biocompatibility and feasibility may pose an issue, therefore the nanoparticle selection will be limited to the following:

This example will limit the wavelength of incoming light to 400 nm to 900 nm, or 7.5×10^{14} Hz to 3.3×10^{14} Hz. The step size will be 50 nm (6×10^{13} Hz). The medium refractive index (RI) for acquiring the optical cross sections will be 1.38 . The selection of the nanoparticles is spherical nanoparticles made of gold , silver , and copper . There will be three nanoparticles of each material: one of diameter 20 nm, one of diameter 60 nm, and one of diameter 100 nm. Both the absorption and scattering cross-sections were given the same weight of 0.5.

Optical cross-sections for this example were generated using the Mie Theory as well, however the Rayleigh theory would have been better for nanoparticle-frequency pairings where the particle diameter was 1/10th the wavelength size.

When the algorithm was run, it identified the optimal combination of three nanoparticle-frequency pairings: gold nanoparticles with a diameter of 100 nm, interacting with light at a wavelength of 600 nm; silver nanoparticles with a diameter of 20 nm, interacting with 900 nm light; and silver nanoparticles of 60 nm, interacting with 450 nm light. These combinations resulted in a total S-score of 5.364865.

Parameter overview: In the above example, biocompatibility and feasibility were considered while utilizing the algorithm however, those are not the only factors to consider. When considering the integration of the described optimization algorithm for nanoparticle control into practical applications, several factors need to be evaluated to ensure feasibility and effectiveness. These factors include biocompatibility, feasibility, material stability, and resource limitations.

Discussion

The optimization algorithm presented in this study effectively reduces crosstalk by maximizing the selectivity of nanoparticle-frequency pairings. By scoring each pairing based on their optical cross-sections, the algorithm ensures that each nanoparticle responds predominantly to the intended frequency, thereby minimizing unintended interactions. This approach can significantly improve independent control over nanoparticles compared to existing methods. Traditional techniques like magnetic, chemical, and mechanical controls often face limitations such as thermal effects, chemical interference, and complex setups. In contrast, the proposed algorithm leverages electromagnetic interactions, particularly localized surface plasmon (LSP) excitations, to fine-tune optical properties and achieve precise control. Utilizing this algorithm in conjunction with LSP-based control significantly increases the potential for procedures on a nano level alongside the implications. The application of frequency-selective nanoparticles holds substantial potential in various fields. As previously mentioned in the example, one area where nanoparticles show great promise is in the field of chemotherapy, which could allow for greater control over the area undergoing treatment, therefore keeping normal cells healthy, which has the potential to save many lives. While the algorithm shows promise, it is important to acknowledge its limitations. The algorithm requires the optical cross-sections for all the nanoparticles with interactions of frequencies representative of the frequency range for the wavelengths. This puts the algorithm at the same limits as its constituent data, and for many materials and shapes, the optical cross-sections are calculated using theoretical approximations.

Conclusion

The paper successfully demonstrates an optimization algorithm designed to enhance frequency selectivity for nanoparticle control, significantly reducing crosstalk. This is

achieved by analyzing extinction, scattering, and absorption cross-sections for various nanoparticle-frequency pairings. The algorithm effectively ensures that each nanoparticle responds predominantly to the intended frequency, minimizing unintended interactions and maximizing control precision. These findings underscore the potential of using electromagnetic interactions, particularly localized surface plasmon (LSP) excitations, to fine-tune optical properties for precise control.

This method can significantly impact technology by advancing the design and functionality of nanodevices and enabling more precise and independent control. Such precise control is crucial in various fields, including targeted drug delivery, where frequency-selective nanoparticles can ensure localized chemotherapy with minimal systemic exposure and enhanced therapeutic efficacy. Similarly, in nanoelectronics, the ability to control nanoparticles independently enables the development of more efficient and customizable devices. This approach promises substantial improvements in medical diagnostics, drug delivery, and the broader sphere of nanoelectronics.

I recommend experimental validation to confirm the simulation results, addressing the limitation of relying solely on theoretical simulations. Future studies should also explore a wider range of variables, such as non-spherical shapes and other materials, to further optimize the algorithm. This could involve investigating the stability of multi-layered nanoparticles and the chemical properties required for practical implementation. Such directions are vital as they could uncover potential issues and refine the algorithm for broader and more practical applications.